

Space shuttle's impact on the stratosphere: An update

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Abstract. To assess their impact on the stratosphere, a launch scenario of nine shuttles and three Titans per year is simulated in a two-dimensional photochemistry and transport model that includes heterogeneous reactions on a stratospheric sulfate aerosol (SSA) layer and polar stratospheric clouds (PSCs). These rocket launches are predicted to cause small constituent changes in the stratosphere. Maximum total inorganic chlorine enhancements are computed to be about 12 parts per trillion by volume ($\sim 0.4\%$ on a 3 parts per billion by volume background) in the middle to upper stratosphere at northern middle to high latitudes. Maximum ozone decreases associated with these chlorine increases are calculated to be about 0.14% in the middle to upper stratosphere at northern middle to high latitudes. Column ozone decreases are predicted to be a maximum of about 0.05% at northern polar latitudes in the early spring. Model results using (1) gas phase only reactions, (2) gas phase reactions and heterogeneous reactions on the SSA layer, and (3) gas phase reactions and heterogeneous reactions on the SSA layer and PSCs have also been compared with one another. The simulations from these three versions of our model gave annually averaged global total ozone decreases of (1) 0.0056%, (2) 0.010%, and (3) 0.014%. Stratospheric effects from heterogeneous reactions promoted by the alumina emitted from these rockets could be larger than those predicted from the chlorine emissions and need to be investigated further.

1. Introduction

The launch of NASA's space shuttle and similar rockets inject hydrogen chloride, carbon monoxide, water vapor, molecular nitrogen, and aluminum oxide directly into the stratosphere. The global effects of the chlorine compounds emitted by the solid rocket boosters on the stratospheric ozone layer have been studied previously [Prather *et al.*, 1990, hereinafter referred to as PR90] with the use of models which included only gas phase chemical reactions. The purpose of this study is to update the PR90 work using a global two-dimensional model that includes heterogeneous chemical reactions on the background stratospheric sulfate aerosol (SSA) layer and polar stratospheric clouds (PSCs).

Hydrolysis of N_2O_5 on the SSA layer changes the partitioning in the lower stratosphere for the odd nitrogen family by converting NO_x constituents (NO , NO_2 , NO_3 , N_2O_5) to HNO_3 . This repartitioning of odd nitrogen constituents in the lower stratosphere increases the importance of the HO_x (H , OH , HO_2) and Cl_x (Cl , ClO , Cl_2O_2) cycles for ozone destruction at the expense of the NO_x cycles [Rodriguez *et al.*, 1991]. The inclusion of heterogeneous reactions on the SSA layer in atmospheric models also leads to better agreement between the model predictions and measurements of odd nitrogen constituents [Prather and Remsberg, 1993]. In addition, heterogeneous reactions on PSCs play a critical role in chlorine activation and subsequent ozone depletion in the polar regions [Solomon, 1988]. Since both the SSA layer and the PSCs have been shown to enhance ozone depletion re-

sulting from chlorine compounds [e.g., Chipperfield and Pyle, 1988; Solomon, 1988; Hofmann and Solomon, 1989; Isaksen *et al.*, 1990; Rodriguez *et al.*, 1991; Brasseur and Granier, 1992; Considine *et al.*, 1994], it was expected that predictions of larger ozone depletions from the solid rocket booster emissions would result from the simulations reported below compared with those computed by PR90.

The potential damage to stratospheric ozone by the chlorine released from solid-fuel rockets was first recognized two decades ago as part of the Climatic Impact Assessment Program [Hoshizaki, 1975]. Potter [1978] computed total ozone losses of 0.25% in the northern hemisphere if a launch rate of 60 space shuttles per year were maintained. Since 1990, several studies have been completed using more reasonable launch rates and updated chemistry to study the stratospheric impact of rockets on local, regional, and global geographic scales. The global scale effects of the rocket launches are best understood within the context of the local and regional effects, especially given the expectation that these local and regional effects on stratospheric constituents are more substantial than the global effects.

A brief overview of the previous work on the stratospheric effects of rocket launches is given in section 2. The two-dimensional photochemical and transport model simulations are described in section 3 and the constituent results of these model simulations are discussed in section 4.

2. Overview of Stratospheric Effects From Rocket Launches

Local effects of rocket launches (in the exhaust plume or close by) have been studied by Karol *et al.* [1992], Kruger *et al.* [1992], Danilin [1993], Denison *et al.* [1994], and Kruger [1994].

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Ozone in the plume is predicted to be decreased by more than 80% in both the *Karol et al.* [1992] and the *Denison et al.* [1994] computations. These huge ozone depletions are predicted to last from several minutes [Denison et al., 1994] to several hours [Karol et al., 1992]. Pergament et al. [1977] show a measurement of ozone reduction of more than 40% below background in the exhaust trail of a Titan 3 solid rocket at an altitude of 18 km at a time of 13 min after launch. Since this measurement has only been performed once and is of uncertain reliability (M. Loewenstein, private communication, NASA Ames Research Center, 1991), it is unclear if the substantial ozone losses predicted by models for the exhaust plume have been validated or not.

Kruger et al. [1992] conducted a range of simulations for a variety of initial perturbations of HCl. Both Danilin [1993] and Kruger [1994] investigated the local effects among a large range of possible perturbations of HCl and Cl₂ by the rockets and found that the local ozone response was found to be very sensitive to the Cl₂ content of the exhaust. Danilin [1993] also included heterogeneous reactions that might proceed on the Al₂O₃ particle surfaces from the rocket exhaust and found that the considered heterogeneous reactions did not play an important role in the ozone response for the exhaust plume.

Aftergood [1991] speculated that there could be a significant local decrease in column ozone after a space shuttle launch. Computations by Karol et al. [1992], Danilin [1993], and Kruger [1994] indicate that the maximum depletion of the column ozone over any one point is less than 10% since the space shuttle trajectories are curved rather than vertical. Consistent with these model computations, McPeters et al. [1991] did not find any evidence of ozone depletion in a study of total ozone mapping spectrometer (TOMS) data taken at varying times after eight shuttle launches.

Both PR90 and Hirschberg [1993] applied three-dimensional models to compute the regional atmospheric effects due to single Space Shuttle launches. PR90 found peak enhancements of about 30 pptv in total inorganic chlorine, Cl_y (Cl, ClO, ClONO₂, HOCl, and HCl), over a 20° latitude by 30° longitude region two days after a launch. Hirschberg [1993] found the maximum O₃ depletion associated with the Cl_y increase to be ~0.4% occurring about 3½ days after launch at 35.7 km.

PR90 showed that the Cl_y emitted by the shuttle becomes spread over all longitudes in about 30 days, suggesting that studies concentrating on the cumulative long-term global-scale effects can be done reasonably well with the use of zonal-mean (two dimensional) models. Global influences have been computed with two-dimensional models for space shuttle and Titan 4 launches in PR90, Ariane 5 launches in the works of Pyle and Jones [1991] and Jones et al. [1995], SANGER spacecraft flights in the work of Bruhl [1993], National Aerospace Plane (NASP) and SANGER spacecraft flights in the work of Jackman et al. [1992], and several different space launch systems in the work of Kolb et al. [1990]. The SANGER spacecraft and NASP are future space transport systems. Recent summaries of the effects of exhaust products from rockets are given in the works of World Meteorological Organization (WMO) [1991], American Institute of Aeronautics and Astronautics (AIAA) [1991], and Jackman [1994].

3. Two-Dimensional Model Simulations

PR90 assessed the steady state impact of nine space shuttles and six Titan 4 launches per year on the chlorine loading and ozone abundance with two two-dimensional models that in-

Table 1. Stratospheric Chlorine Released Annually by the Space Shuttle and Titan 4 Launches

Altitude, km	Kilograms of Chlorine (as Cl)		
	A	B	C
15–20	176,800	16,800	8,400
20–25	136,100	14,700	7,300
25–30	109,600	12,600	6,300
30–35	87,400	10,600	5,300
35–40	69,300	8,900	4,500
40–45	25,900	7,300	3,700
45–50	4,500	6,000	3,000

From Table 1, Prather et al. [1990]. The launch scenario assumes nine shuttles (A) and two Titans (B) from Cape Canaveral, Florida (29°N, 80°W), and one Titan (C) from Vandenberg AFB, California (34°N, 121°W).

cluded gas phase only chemistry. Since PR90 was published, an error of a factor of 2 was discovered in the Titan 4 chlorine release rate/launch [Brady et al., 1994]. The actual scenario simulated by PR90 was for nine space shuttles and three Titan 4 launches, fairly close to the historical launch rates for the 1990s [Brady et al., 1994]. We use the same launch rate scenario described by PR90 in this study for two reasons: (1) The rate of launches in PR90 is fortuitously close to historical launch rates and (2) comparisons with the PR90 results are more straightforward. The assumed stratospheric chlorine released annually is given in Table 1.

PR90 estimated the annual source of stratospheric chlorine from man-made industrial halocarbons to be about 300 kilotons of chlorine per year for 1990 conditions. This is much larger than the 725,000 kg (0.725 kt) of total chlorine released per year into the stratosphere by the solid rocket motors above 15 km computed from Table 1. Our assumed launch rate would add only another 0.25% to the stratospheric chlorine accumulating from other anthropogenic activities (see PR90 for further discussion). Since industrial sources of stratospheric chlorine are expected to decrease because of the internationally agreed on regulations [WMO, 1994], it is expected that the fractional contribution from rocket emissions would become larger if the launch rate remains constant or increases.

Six different model simulations are studied here: (A) a base simulation (no rocket launches) including gas phase only reactions, called “gas only”; (B) “gas only” with the rocket launches; (C) a base simulation (no rocket launches) including the reactions on the SSA layer along with the gas phase reactions, called “Het”; (D) “Het” with the rocket launches; (E) a base simulation (no rocket launches) including the reactions on the SSA layer and PSCs along with the gas phase reactions, called “Het and PSCs”; and (F) “Het and PSCs” with the rocket launches. These six model simulations are best considered as three pairs of simulations: a base simulation with no rocket launches and a perturbed simulation with the rocket launches. In this way, simulations A and B, C and D, and E and F are paired together.

All simulations were completed with the latest version of the Goddard Space Flight Center (GSFC) two-dimensional photochemical and transport model which is the result of several years of development. The model was first described by Douglass et al. [1989] and its altitude range was extended through the mesosphere in the work of Jackman et al. [1990]. Reaction rates in the model have been updated to DeMore et al. [1994].

Table 2. Ground Boundary Conditions for Source Gases Approximating the Steady State Situation for Year 1990

Source Gas	Value (Mixing Ratio)
N ₂ O	308 ppbv
CH ₄	1.683 ppmv
H ₂	500 ppbv
CO	150 ppbv (NH)
CO	50 ppbv (SH)
CH ₃ Cl	600 pptv
CFCl ₃	246 pptv
CF ₂ Cl ₂	422 pptv
CCl ₄	105 pptv
CH ₃ CCl ₃	136 pptv
CHClF ₂	88 pptv
C ₂ F ₃ Cl ₃	55 pptv
C ₂ F ₄ Cl ₂	7 pptv
C ₂ F ₅ Cl	5 pptv
CH ₃ Br	14 pptv
CBrClF ₂	2.3 pptv
CBrF ₃	1.4 pptv

Adapted from WMO [1995]. NH, northern hemisphere; SH, southern hemisphere; ppbv, parts per billion by volume; ppmv, parts per million by volume; pptv, parts per trillion by volume.

Heterogeneous reactions on the PSCs were included with a parameterization described by *Considine et al.* [1994]. A lookup table formulation is now used to compute the photolytic source term over the model grid and is discussed by *Stolarski et al.* [1995, Appendix A under the GSFC section]. The model has a “fixed dynamics” formulation based on National Meteorological Center temperature data with heating rates computed off line and the horizontal eddy diffusion coefficients (K_{yy} values) calculated to be consistent with the residual circulation [see *Fleming et al.*, 1995]. The model’s temperatures, residual circulation, and K_{yy} values change monthly but repeat yearly. The “fixed dynamics” allow us the ability to compare easily the photochemical changes among the simulations of “gas only,” “Het,” and “Het and PSCs” without the complication of transport changes among the three simulations.

All model simulations were run for 20 model years to a seasonally repeating steady state condition. The ground boundary conditions of the long-lived source gases for the steady state 1990 conditions are given in Table 2. These boundary conditions are used in all simulations and input about 3.3 parts per billion by volume (ppbv) organic chlorine at the ground. The perturbed runs with the rocket launches (B, D, and F) were then compared to those base runs without any rocket launches (A, C, and E).

Solid rockets are known to emit carbon monoxide, water vapor, molecular nitrogen, aluminum oxide, and other constituents in minor amounts, as well as hydrogen chloride, directly into the stratosphere [AIAA, 1991; WMO, 1991]. To date, most studies have focused on the chlorine compounds emitted by the solid rockets as the largest threat to stratospheric ozone. This assumption is reasonably justified because the fraction of ozone loss is expected to be the largest for the chlorine compounds than for any of the other emitted chemicals. *McDonald et al.* [1991] have shown that the relative percentage of stratospheric ozone loss is increased due to solid rockets by the following amounts: NO_x by $5 \times 10^{-5}\%$, HO_x by $1.2 \times 10^{-3}\%$, and Cl_x by $3.2 \times 10^{-2}\%$. These computations indicate that solid rocket motors with ammonium perchlorate as the oxi-

dizer influence ozone mainly through the chlorine catalytic cycles. For these reasons we only include the hydrogen chloride emissions in the model computations presented here.

We have not considered the effects of solid rocket emissions of Al₂O₃ in our model studies. However, the effects of emitted Al₂O₃ may need to be considered more carefully in a future study, because they add to the background particle burden and to the stratospheric heterogeneous processes. *PR90 and McDonald et al.* [1991] roughly estimate that the launch rate assumed in this study would increase the global aerosol surface area by about 0.1% from the Al₂O₃ being emitted by the rockets. If these Al₂O₃ (alumina) particles are coated with sulfate, then they would act in a similar way to the SSA layer. An increase of 0.1% in the SSA layer would increase the predicted ozone loss associated with the “Het” cases by only about another 1/1000th, an insignificant enhancement. On the other hand, if the Al₂O₃ particles are not coated with sulfate, then a more substantial ozone loss may occur. *Meads et al.* [1994] measure a reaction probability of the ClONO₂ + HCl → Cl₂ + HNO₃ process to be 0.01–0.02 on an α-alumina surface. Although this reaction is a factor 15–30 smaller than the accepted reaction probability for the ClONO₂ + HCl process on PSCs, it still could be important. The alumina particles would be spread over the entire stratosphere over the entire year, whereas the PSCs are produced only at the polar latitudes in the lower stratosphere in winter. Presently, not enough is known about the alumina emitted by the solid rockets to complete a more detailed analysis of the problem.

A recent report [Beiting, 1995] summarizes the state of knowledge of alumina ejected by launch vehicles. *Cofer et al.* [1984] found that over 72% of the exhausted alumina was α-alumina. Conversely, *Dill et al.* [1990] have concluded that more than 64% of the solid propellant exhaust consists of γ-alumina. Three key questions that must be answered before the effects of alumina can be evaluated are: (1) Will the emitted alumina become coated by sulfate?, (2) What is the proportion of alumina in the exhaust between α-alumina and γ-alumina?, and (3) Do exhausted γ-alumina particles promote the ClONO₂ + HCl → Cl₂ + HNO₃ reaction?

4. Two-Dimensional Model Constituent Results

The results of the model’s 20-year simulations for the “gas only,” “Het,” and “Het and PSCs” studies are presented in this section. The model’s numerical precision for these simulations is <0.001%. Thus our model predictions are repeatable for atmospheric perturbations which are a few times greater than 0.001% and are out of the model’s numerical noise. Although the rocket emissions cause fairly small perturbations to the atmosphere, as discussed below, they are large enough so that our model’s precision is adequate.

The computed Cl_y increases are given in Figures 1a and 1b for January and July, respectively. These Cl_y increases do not change significantly among the three pairs of simulations. This is because the source of Cl_y from the rocket exhaust is the same in all cases, the model’s dynamics were kept the same, and the simulated ozone changes were not large enough to substantially change the stratospheric photochemical environment. Maximum chlorine enhancements are computed to be about 12 pptv (~0.4% on a 3 ppbv background) in the middle to upper stratosphere at northern middle to high latitudes for both January and July. These computed maximum chlorine enhancements are quite similar to those presented in Figures

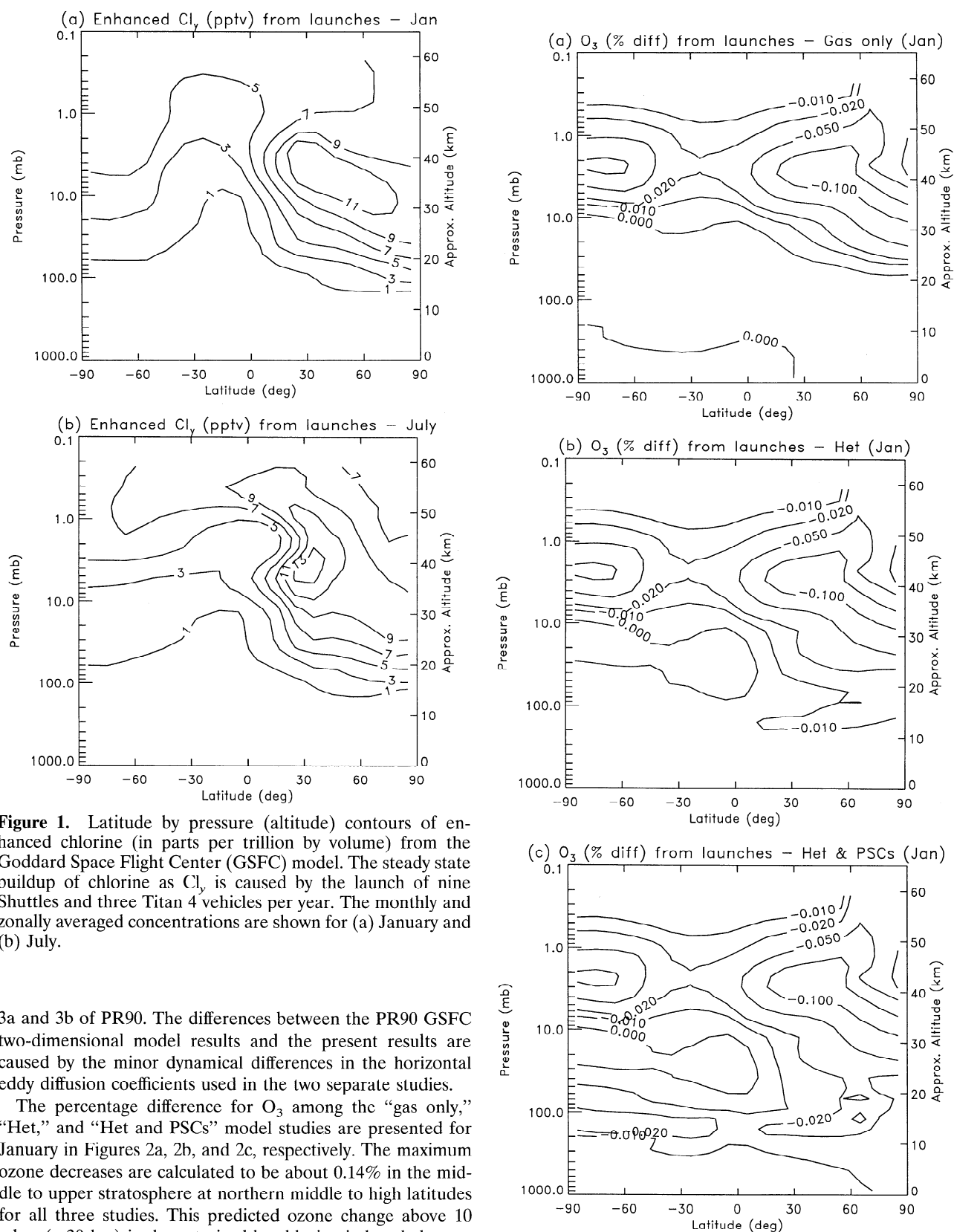


Figure 1. Latitude by pressure (altitude) contours of enhanced chlorine (in parts per trillion by volume) from the Goddard Space Flight Center (GSFC) model. The steady state buildup of chlorine as Cl_y is caused by the launch of nine Shuttles and three Titan 4 vehicles per year. The monthly and zonally averaged concentrations are shown for (a) January and (b) July.

3a and 3b of PR90. The differences between the PR90 GSFC two-dimensional model results and the present results are caused by the minor dynamical differences in the horizontal eddy diffusion coefficients used in the two separate studies.

The percentage difference for O_3 among the “gas only,” “Het,” and “Het and PSCs” model studies are presented for January in Figures 2a, 2b, and 2c, respectively. The maximum ozone decreases are calculated to be about 0.14% in the middle to upper stratosphere at northern middle to high latitudes for all three studies. This predicted ozone change above 10 mbar (~ 30 km) is characterized by chlorine-induced changes from gas phase chemistry and does not vary much among the three cases. The “Het” case shows an increase in computed ozone loss, especially in the northern middle latitudes between 20 and 30 km. The “Het and PSCs” shows an even more apparent increase in computed ozone loss in the lower stratosphere for both the northern and the southern hemispheres.

Figure 2. Latitude by pressure (altitude) contours of the perturbation to ozone (percent) from the GSFC model caused by the launch of nine shuttles and three Titan 4 vehicles per year. The monthly, zonally averaged computed O_3 changes for January are shown for model simulations of (a) gas only, (b) Het, and (c) Het and polar stratospheric clouds (PSCs).

The computed maximum ozone decreases for the “gas only” case are also quite similar to those presented in Figure 8a of PR90; however, the predicted losses for the lower stratosphere in the “Het” and “Het and PSCs” cases are a new feature in the simulated ozone loss characteristics.

The computed upper stratospheric ozone depletion is over 0.1% for January in both hemispheres, even though the Cl_y increase is clearly higher in the northern hemisphere. Although puzzling at first, this prediction is easily explained. The effect on ozone by chlorine in the upper stratosphere is maximum in the summer, thus the ozone depletion is enhanced in the southern hemisphere and diminished in the northern hemisphere in January. For July, our predictions indicate substantially more northern than southern hemispheric ozone depletion.

Jones *et al.* [1995] also used a two-dimensional model to assess the impact of the chlorine from ten Ariane 5 launches per year. They found chlorine enhancements (maximum of about 0.2%) and ozone depletions (maximum of about 0.09%) in the upper stratosphere, less than but similar in magnitude to these and the PR90 computations.

The ozone decreases simulated in the GSFC two-dimensional model are seasonally dependent and the computed total ozone decreases are given in Figures 3a, 3b, and 3c for the “gas only,” “Het,” and “Het and PSCs” cases, respectively. The maximum column-integrated (total) ozone decreases are predicted to be at northern polar latitudes in the late winter and early springtime period for the “gas only” and “Het” cases. The largest total ozone decreases are predicted to be about 0.05% in the “Het and PSCs” case at polar latitudes in the early spring of both hemispheres. PR90 do not present contour figures of predicted total ozone change, however, they do indicate that the simulated total column ozone loss would be less than 0.1%, a result not contradicted in this study.

Comparison of Figures 2b with 2c and 3b with 3c shows that the low and midlatitude response of model O_3 levels to space shuttle emission is quite different when PSC processes are included in the model formulation. This may at first seem surprising because PSCs occur at the poles and it is natural to expect that their effects would be confined to the polar regions. This is not actually the case. The denitrification, dehydration, and ozone depletion caused by the PSCs are transported to lower latitudes and results in a lower stratosphere in which chlorine chemistry is more important than occurs in the “gas only” and “Het” cases.

It is interesting to compare the predicted launch-induced ozone change in the “Het and PSCs” case to other anthropogenic ozone changes computed with our model by Considine *et al.* [1994]. The ozone changes predicted for halocarbon emissions and emissions from a proposed fleet of stratospheric aircraft are much larger than those resulting from the launches. Anthropogenic halocarbon emissions are predicted to have caused total ozone decreases of about 1% near the tropics to over 15% in the southern polar springtime between 1980 and 1990 [Considine *et al.*, 1994, Figure 8a]. The aircraft fleet of 500 planes flying between 17 and 20 km emitting NO_x at a rate of 15 g/kg of fuel burned in a 1980 background chlorine level condition is predicted to cause maximum total ozone increases of about 0.2% in the northern polar late spring/early summer and maximum total ozone decreases of over 2% in the southern polar springtime [Considine *et al.*, 1994, Figure 10b]. The halocarbon emissions are predicted to cause significantly more ozone change than the HSCT fleet

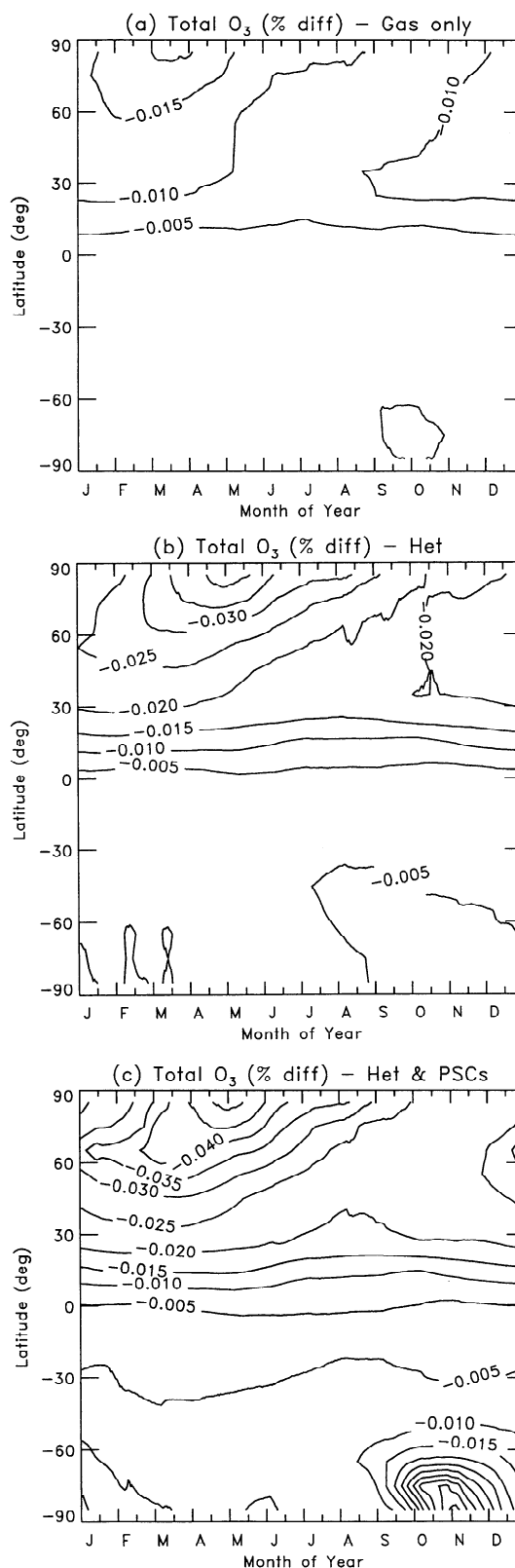


Figure 3. Latitude by month of year contours of the perturbation to total ozone (percent) from the GSFC model caused by the launch of nine shuttles and three Titan 4 vehicles per year. The computed total O_3 changes are shown for model simulations of (a) gas only, (b) Het, and (c) Het and PSCs.

and about 2 orders of magnitude more ozone loss than the launch emissions. The HSCT fleet is predicted, on average, to cause about an order of magnitude more ozone change than the launch emissions.

We can also compare the launch-induced ozone change to the total ozone trends derived from TOMS measurements for the 1979–1990 period [Stolarski *et al.*, 1991]. These total ozone decreases are derived to be about 4–15% in the northern polar latitudes and about 4–30% in the southern polar latitudes. The largest predicted total ozone decrease at polar latitudes from our assumed launch rate scenario ($\sim 0.05\%$) is about 2 orders of magnitude less than that derived from TOMS measurements. Smaller trends are observed in middle latitudes (about 2–8% in both hemispheres) and statistically insignificant trends are observed for the tropics from TOMS measurements. Our assumed launch rate scenario is predicted to cause an ozone decrease of 0.02–0.04% and 0.005–0.015% in northern and southern middle latitudes, respectively, and 0.005–0.025% in the tropics. Our predicted launch-induced ozone decreases are, in general, third-order effects compared to these total ozone trends derived from observations.

The annually averaged global total simulated ozone loss was computed to be 0.0056% for the “gas only,” 0.010% for the “Het,” and 0.014% for the “Het and PSCs” cases. The total predicted ozone loss from a launch rate of nine space shuttles and three Titan 4 rockets per year is thus increased by a factor of about 2.5 when heterogeneous chemistry on the SSA layer and PSCs is considered in the GSFC model. Karol *et al.* [1992] also computed a total global ozone depletion from space shuttle launches. Scaling of the Karol *et al.* [1992] calculations to the launch rate used in this study give losses of 0.0072 to 0.024%, close to present model computations.

Although the updated calculations show a factor of 2.5 increase in shuttle-induced global ozone depletion when heterogeneous chemistry on the SSA layer and PSCs are included, the calculated impact of the present launch rate of space shuttle and Titan 4 rockets is still quite small. Note that these calculations do not consider the effects of heterogeneous reactions on alumina particles emitted by the solid rocket boosters, which could substantially alter the results. Further work characterizing the behavior of alumina particles in the stratosphere is necessary before an evaluation of their effects on global ozone levels can be made.

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